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Dense Membranes for Anode Supported all Perovskite IT-SOFCs

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The intent of the project is to develop inexpensive oxygen permeable, dense and high surface area membranes in the form of bulk and highly oriented thin films using soft solution chemical routes and pulsed laser deposition (PLD) techniques for fabricating natural gas fuelled anode supported all perovskite based intermediate temperature **(IT)-SOFCs** with planar architecture.

This project was initiated at Southern University during the academic year 2003-2004. Since January 2004, a full time research associate, and three students (two graduate and one undergraduate) are working in this project. Several mixed ionic and electronic conducting perovskites were synthesized using wet chemical methods, and investigated their use as oxygen separation membranes for the partial oxidation (PO_X) of methane to syngas.

There are two major obstacles that have to be solved to operate SOFCs at intermediate temperatures, including the performance of electrolyte and electrodes. Lowering the operating temperature is possible with the use of alternative materials, appropriate cell design and manufacturing routes. In the search for dense electrolyte materials, the perovskite based systems (ABO_3) have been considered as alternative options, particularly because ABO_3 can take on a number of different structures, and can be doped with aliovalent cations on both the A and B sites. They can also accommodate very large concentrations of anion vacancies into their structures. $LaGaO_3$-based perovskite type oxides, in particular, Sr- and Mg-doped $LaGaO_3$ (LSGM), exhibit high oxide ion conductivity. The exceptional structural and chemical compatibility of LSGM with $La_{0.9}Sr_{0.1}Co_{0.9}M_{0.1}O_3$ [M= Fe, Ni, Mn] as perovskite-based cathode, and Ni based perovskite cermet as anode, makes it a unique electrolyte for all perovskite based IT-SOFC.

There is a critical need to optimize the processing conditions to obtain well sintered LSGM electrolytes at low temperatures for developing miniaturized SOFC cell and stacks working at 600-800 ^{0}C. The single phase LSGM with high sintered density is not easy to obtain by conventional solid state technique. One of the requisites for application as SOFC electrolytes is high sinterability. The extent of sintering depends on the mode of synthesis. The solid state route results in hard agglomerates and coarser grains which inhibit sintering to obtain dense electrolyte materials. In view of increasing importance to produce dense LSGM ceramics on a large scale with better phase purity at lower temperatures in a cost effective manner for use as electrolytes, an investigation was carried out to study the effect of conventional and microwave assisted sintering of the recycled LSGM samples obtained from RSG route which is a combination of solid state reaction and Pechini-type method. The ability to recycle the same sample and use

different processing conditions to tailor the properties of LSGM samples, makes our

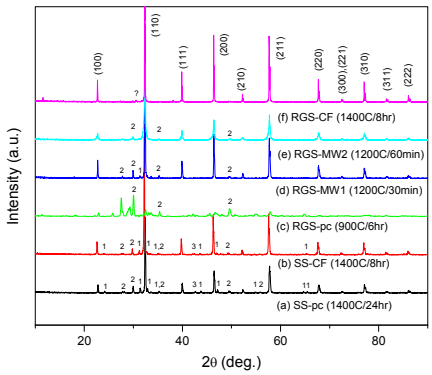

Fig 1. The XRD patterns of $La_{0.8}Sr_{0.2}Ga_{0.85}Mg_{0.15}O_{2.825}$: (a) solid-state route precursor, (b) solid-state route pellet by conventional furnace sintering, (c) regenerative sol-gel (RSG) route precursor calcinated at 900°C for 6 hr, (d) RSG pellet by one-cycle microwave sintering, (e) RSG pellet by two-cycle microwave sintering, and (f) RSG pellet by conventional furnace sintering. Secondary phase peaks are indicated as: "1", $LaSrGaO_4$; "2", $LaSrGa_3O_7$; "3", possible MgO

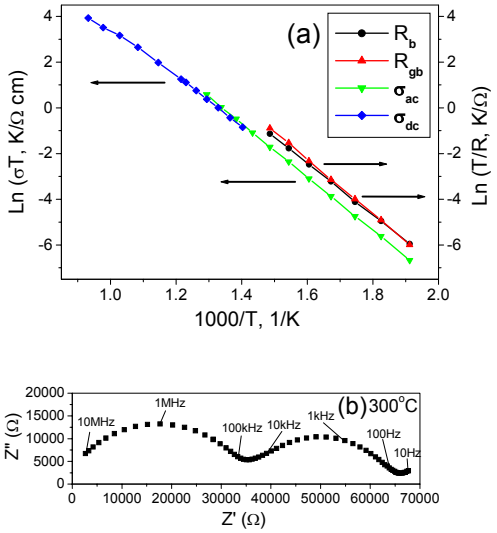

Fig 2 a & b. Conductivity measurement for the regenerative sol-gel $La_{0.8}Sr_{0.2}Ga_{0.85}Mg_{0.15}O_{2.825}$ by conventional furnace sintering: (a) Arrhenius relations for resistance in the bulk (R_b), resistance in the grain-boundary (R_{gb}), conductivity measured by ac method (σ_{ac}), and conductivity measured by dc method (σ_{dc}), (b) complex-plain impedance spectrum at 300ºC.

regenerative sol-gel (RSG) technique innovative and cost effective.

In this presentation, we report a phase pure $La_{(1-x)}Sr_xGa_{(1-y)}Mg_yO_{3-(x+y)/2}$ (LSGM) [(x, y)= (0, 0), (0.1, 0.1), (0.2, 0.15), (0.2, 0.17)] electrolytes, prepared by RSG and sintered at 1200 °C which is lower than the sintering temperatures reported so far. The effect of conventional and microwave heating on sintering, the phase formation, phase purity, and density of samples obtained from both the solid state and regenerative route was investigated. Microwave heating was carried out using SiC as a microwave susceptor. The XRD (Fig 1), SEM, HRTEM, XANES, and ac electrochemical impedance (Fig 2 a & b)measurements (250 -500 °C) were performed. The highly sintered samples exhibited particle size in the range 20-50 nm. The electrical conductivity of these samples was compared with the data obtained from the different compositions of the LSGM system in the literature. The conductivity of LSGM electrolytes are higher at lower working temperature compared to conventionally used YSZ (0.16 S/cm at 1000°C). The RSG technique developed in this work enables recycling of the undesired product and subsequently yield product with much better phase purity and density than that obtained from the solid state route and provides the scope for optimization of processing parameters for obtaining dense LSGM for large scale synthesis.

The nanocrystalline $Ce_{0.9}Gd_{0.1}$ $O_{1.95}$ and lanthanide-doped ceria powders and multilayer thin films (using PLD) were prepared and characterized. These electrolytes show promise with LSGM if they are used together either to block the electronic component in the doped ceria with the LSGM, or with the ceria, to block unwanted anode/electrolyte chemical reactions and/or to provide a catalytic surface for fuel oxidation at the anode. The surface layer on the anode side of these electrolytes can make these electrolytes viable for reduced temperature operations.